Development of residual radioactive contamination in the Ploučnice River basin (Bohemia) due to uranium mining in the period 1992 - 2009

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Abstract. Since the end of the sixties of the last century, both underground mining and in-situ acid leaching have been used to exploit uranium in Stráž pod Ralskem locality in Northern Bohemia. Before putting a Central Decontamination Station into operation in 1989, the Ploučnice River was contaminated by waste waters, which contained radium 226 and uranium. During high flows, sediments and suspended solids from the river polluted also the flood plain areas. The uranium mining was terminated in 1994 and subsequently a programme was established for monitoring of the changes in the basin contamination by radioactive substances. The paper describes selected results and conclusions derived from the monitoring.

1. INTRODUCTION

Since the end of the sixties of the last century, both underground mining and in situ leaching have been used to exploit uranium in Stráž pod Ralskem locality. The underground mining was accompanied by extraction of high amount of mine water (in a rate of about 500 l/s). This water contained high concentrations of radium 226 and uranium, particularly in dissolved form. Before putting a Central Decontamination Station into operation in 1989, the Ploučnice River was contaminated by the waste waters. The uranium mining was terminated in 1994 and the chemical extraction, which did not have impact on water radioactivity in the Ploučnice River, in 1996. Since 1996, the impacted localities have been gradually restored. Since the beginning of the uranium mining until 1999, the hydrosphere was permanently monitored for its contamination by radioactive substances, particularly by radium 226. The main results of the monitoring are described in research reports of T.G.M. Water Research Institute [1], [2], [3], [4]. In July 1981, an extreme flood on the Ploučnice River caused contamination of its flooded area by riverbed sediments. A monitoring system was consequently established to monitor the changes in the hydrosphere contamination in the Ploučnice River basin by radioactive substances, particularly in relation to high flows.

2. METHODS

2.1. Gamma radiation

The monitored cross sections were fixed by using granite blocks (at Hradčany site, the monitoring sites were signed on a bridge). The gamma radiation was monitored between the blocks in the selected river cross sections. The location of the cross sections is shown in Figure 1 (D1 Mimoň and D2 Mimoň (left and right bank), D3 Hradčany inflow and D4 Hradčany outflow, D5 Boreček and D6 Brenná, D7 Žízníkov right bank and D8 Žízníkov left bank). The in situ measurements of gamma radiation were performed at fixed sites of the cross

sections at the height of one meter above the ground. The gamma radiation was measured by using TESLA NB 3201 device during a time period of 10 seconds subsequently for three times in pGy/s and for graphical interpretation the results were converted to μGy/h. The relative standard deviation at 95% level of significance was 15.6% for values around 30 μGy/h, 3.9% for 130 μGy/h and 2.8% for 300 μGy/h.

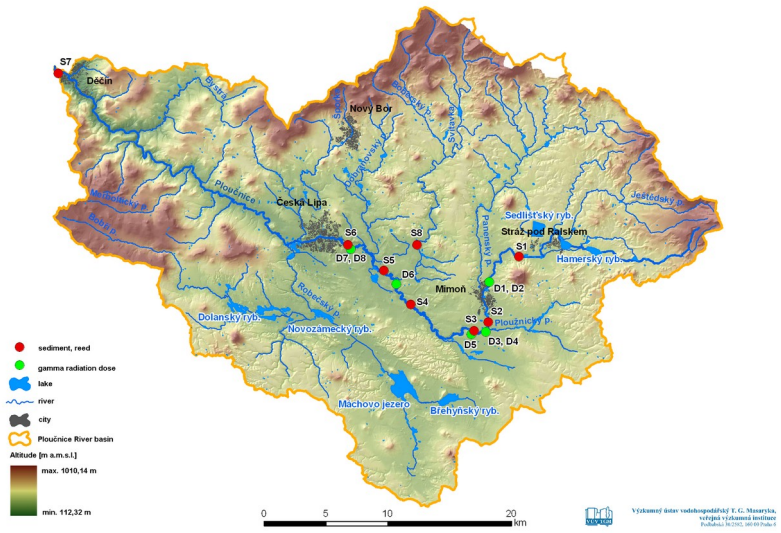

FIG. 1. Location of cross sections for monitoring of gamma radiation (D1 to D8) and sampling of sediments (S1 to S8) in the Ploučnice River basin.

2.2. Sampling and gammaspectrometric analysis of sediments

The sampling was performed at 7 river sites - S1 Noviny, S2 Mimoň laguny, S3 Boreček, S4 Veselí, S5 Vlčí Důl, S6 Žízníkov and S7 Děčín Zámecký rybník. The Svitávka River at Zákupy (S8) was chosen as reference (not contaminated) river site. The location of the S1 to S8 river sites is shown in Figure 1. The sediment samples were taken from the depth of 10 cm below the ground surface and homogenised by their mixing in a plastic box. Same of the samples were used for their grain size analysis performed by ARCADIS Geotechnika a.s. Praha (formerly Geotechnika a.s. Praha) Laboratories [5]. The grain size of all of the samples was below 2 mm. In the Laboratory, the samples were dried at temperature of 105 °C to their constant weight and located into Marinell boxes, whose volume was 450 ml. Activities of the radionuclides emitting gamma radiation were subsequently determined by using methods specified in Standard ČSN ISO 10703 (75 7630) [6] on Water quality – Determination of radionuclide activities by spectrometric measurement of gamma radiation with high resolution.

3. ASSESSMENT OF THE RESULTS AND DISCUSSION

3.1. Gamma radiation in the flooded area

The results of the gamma radiation monitoring were interpreted graphically. An example of the results for a contaminated site (D5 Boreček) is given in Figure 2.

The gamma radiation values from 2009 were compared with minima and maxima from the period 1992-2004. In the contaminated site (D5 Boreček, Figure 2), the results from 2009 in the area of the first maximum at the distance of 6 – 22 m were moderately below the minimum values from the preceding period. In the other observation sites, the gamma radiation values were at the level of lower range or moderately below this level in the whole length of the cross section. The radiation level as specified for landfills in a Decree of the State Office for Nuclear Safety No. 307/2002 [7] of 0.4 μSv/h was exceeded at the distance of 2 – 22 m from the bank of the Ploučnice River. This criterion, originally specified for landfills, was also used for the assessment of the of the gamma radiation in the flooded areas. The observed values in Gy were considered to be identical with the input of dose equivalent in Sv (§ 57, point e of the decree of the State Office for Nuclear Safety [7]), which specifies that deposited material meets the requirement stipulated in point a), it does not cause an increase in the radiation equivalent by more than 0.1 μSv/h

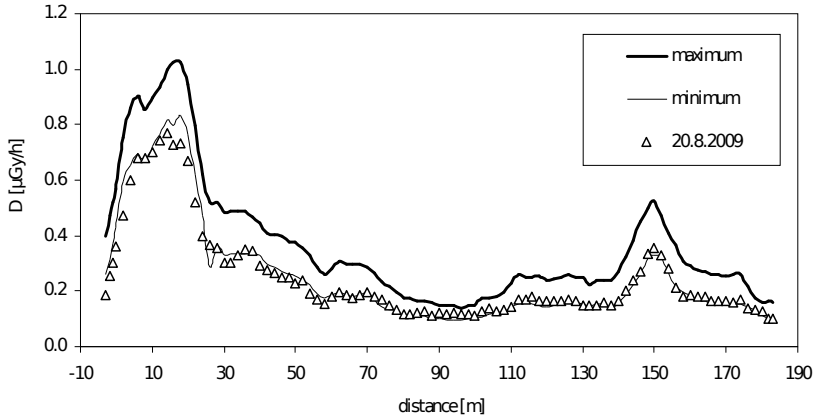

FIG. 2. Gamma radiation in 2009 in an interval of maxima and minima from 1992 – 2004, D5 Boreček cross section.

(as compared to the original value) at the distance of 1 m from the landfill surface and that the input of the dose equivalent does not exceed 0.4 μSv/h.

All of the values of the gamma radiation from 2009 in the uncontaminated site (D8 Žízníkov) were within the minimum and maximum limits from the period 1994 – 2004. The value of 0.1 μSv/h was not exceeded at any observation site in the cross section.

3.2. Gamma radiation in the selected sites of the monitored cross-sections

The data from the period 1994 – 2004 were analysed in terms of their time variability at D4 Hradčany outflow, D5 Boreček and D6 Brenná cross sections. The analysis was performed for two of the observation sites selected at certain distances from zero point in the flooded areas along each of the cross sections. The observed data were interpolated by using logarithmic, linear and exponential function. Coefficients of correlation exhibited the best fit for the logarithmic function at 3 sites, the linear function at 2 sites and the exponential function at 3 sites. The logarithmic function was used in the form of kinetic equation of the first order:

$$\ln D_{i,j} = -\lambda_{ef,i,j} \times t + q_{i,j} \qquad (1)$$

where
- $D_{i,j}$ gamma radiation at selected monitoring site j of cross section i (μGy/h)
- $\lambda_{ef,i,j}$ effective ecological (observed) constant that describes decrease in the radiation at cross section i, site j (1/yr)
- t time (yr)
- $q_{i,j}$ Parameter of the kinetic equation at cross section i, site j

The constant $\lambda_{ef,i,j}$ was used for calculation of the effective ecological half-life by using the equation as follows:

$$T_{ef,i,j} = \frac{\ln 2}{\lambda_{ef,i,j}} \qquad (2)$$

where
- $T_{ef,i,j}$ observed effective ecological half-life of the gamma radiation at cross section i, site j (yr)

For description of the half-life of the radiation component relevant to the former uranium ore mining, the total radiation was corrected by subtracting its background level:

$$D_{i,j}' = D_{i,j} - D_b \qquad (3)$$

where
- $D_{i,j'}$ gamma radiation at selected monitoring site j of cross section i (μGy/h) corrected by using the background radiation D_b, which was substituted by the mean radiation calculated for the Žízníkov uncontaminated site (μGy/h)

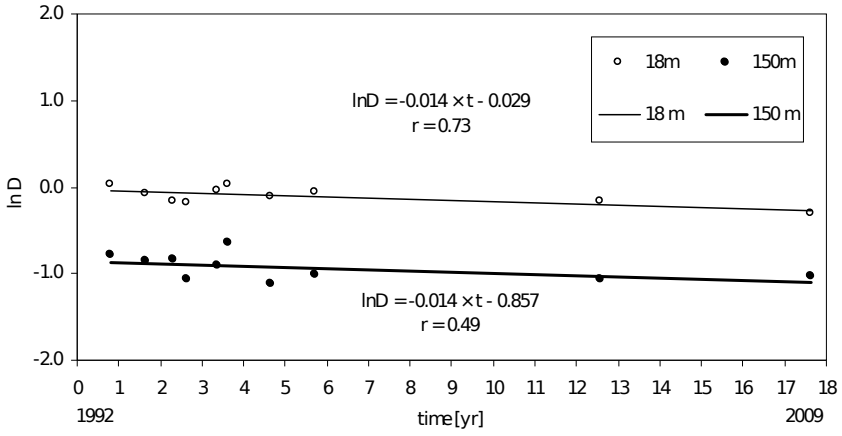

FIG. 3. Time development of gamma radiation (D) at distances of 18 m and 150 m, D5 Boreček cross section, 1992 – 2009.

Table 1. Constants of gamma radiation decrease and effective ecological half-lives before and after subtraction of the background radiation, selected sites of the monitoring cross sections.

Ploučnice River site		l	λ_{ef}	T_{ef}	λ_{ef}'	T_{ef}'
		(m)	(1/r)	(r)	(1/r)	(r)
Mimoň	D1	12	0.019	37.3	0.029	23.6
		22	0.046	15.2	0.057	12.1
Hradčany – inflow	D3	18	0.084	8.2	0.11	6.3
		44	0.085	8.2	0.1	6.9
Hradčany – outflow	D4	22	0.041	16.9	0.059	11.7
		38	0.045	15.3	0.056	12.3
Boreček	D5	18	0.014	49.2	0.015	45.6
		150	0.014	49.5	0.017	42
Brenná	D6	60	0.016	42.3	0.042	16.6
		120	0.08	8.7	0.108	6.4

The corrected constant $\lambda_{ef,,i,j}'$ and effective ecological half-life $T_{ef,i,j}'$ were calculated by substituting $D_{i,j}'$ for $D_{i,j}$ in Equation (1). Figure 3 shows an example of time changes in the gamma radiation, which is interpolated by using logarithmic function. The coefficients of correlation r of the decrease interpolated by the logarithmic function were in the range 0.49 – 0.95, by the linear function in 0.48 – 0.96 and by the exponential function in 0.56 – 0.84. Analogically, for the corrected values of the gamma radiation, the correlation coefficients were 0.49 – 0.94, 0.48 – 0.96 and 0.57 – 0.81.

Table 1 gives values of the effective ecological constants, effective ecological half-times of the gamma radiation and their corrected values. The values of λ_{ef} and Tef substantiate that the gamma radiation (contamination) was decreasing most rapidly in D3 Hradčany inflow and D4 Hradčany outflow cross sections, where the effective ecological half-life was 8.2 yr. D5 Boreček cross section exhibited the smallest decrease in the gamma radiation with effective ecological half-lives of 49.2 yr and 49.5 yr. The corrected half-lives were decreasing more slowly as compared to the uncorrected values. The half-lives in the identical cross sections were 6.3 yr (6.9 yr), 45.6 and 42 yr.

3.3. Contamination of river sediments by radioactive substances

The results of the monitoring of radium 226 activities in bottom sediments that were sampled in a network of cross sections S1 to S8 in the period 1994 – 2009 are shown in Figure 4. The range of the radium 226 activities in bottom sediments from the cross sections of the Ploučnice River was relatively wide. Certain variability is probably attributable to differences in grain size between the individual cross sections and other factors associated with sampling. The results show that the activities in S1 cross section, which is located closely

downstream from the waste water outflow from uranium ore mining at Stráž pod Ralskem, were relatively small as compared to those at the remaining cross sections. This fact can be explained by the highest grain sizes of the sediments from this section. The graphical interpretation of the results suggests that the sediments in upstream reaches were transported and subsequently deposited in the area between S2 Mimoň and S5 Vlčí Důl cross sections. During the whole observation period, the highest activities of radium 226 were detected in sediment samples from S3 Boreček cross section. The lowest values were detected for S1 Noviny and S6 Žízníkov cross sections. It could be generally concluded that the radium 226 activities were significantly decreasing during the whole monitoring period 1994 – 2009.

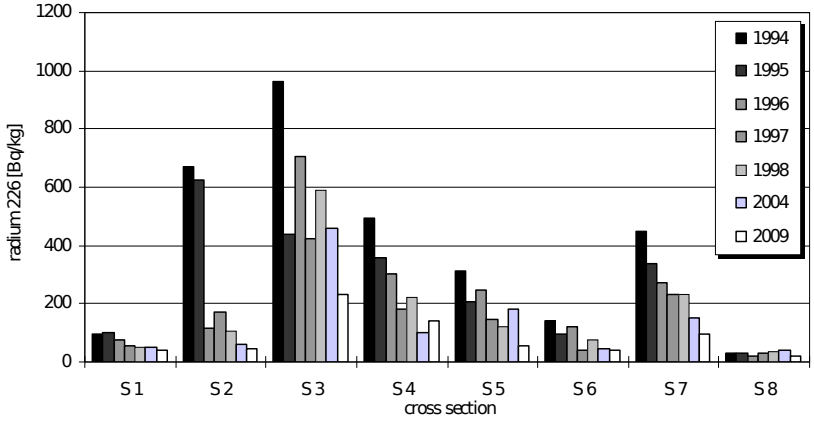

FIG. 4. Radium 226 activities in bottom sediments from the Ploučnice River (S1 to S7 cross sections) and from the uncontaminated cross section of the Svitávka River (S8) in the period 1994 – 2009.

The analysis was also focused on a ratio of the activities of radium 226 and radium 228 in the sediment samples. The results could be considered to be an indicator of the impact of uranium ore mining and processing. The mean value of the ratio in the contaminated cross sections S1 to S7 on the Ploučnice River was 11.9 in 1994, 7.8 in 1995, 6.2 in 1996, 5.9 in 1997, 5.3 in 1998, 4.7 in 2004 and 4.2 in 2009. The mean value of this ratio in uncontaminated river sediments in the Czech Republic is 1.0. For the uncontaminated cross section, the S8 Svitávka Zákupy, the mean ratios in the specified years were 1.2, 1.6, 1.1, 1.4, 1.5, 1.5 and 1.6, with the mean value of 1.4. The mean value is relevant to the first class specified for contamination of river sediments by radium 226 [8], [9]. The range of the ratio at the S8 reference locality can be attributed to uncertainties in determination of low values of radium 226 and 228.

The results of previous studies, which were focused on determination of this ratio at localities that were used for land-filling of waste from uranium ore mining and processing [9] show that the ratio indicates contamination of sediments if it exceeds 1.5. This ratio can also substantiate the decrease in the sediment contamination by radium 226 along the Ploučnice River.

4. CONCLUSIONS AND RECOMMENDATIONS

The paper assesses changes in the contamination of the flooded area by radioactive substances by using the results of gamma radiation monitoring and those from the monitoring of radium 226 and radium 228 activities in bottom

sediments sampled at selected cross sections of the Ploučnice River during the period 1992 – 2009. The results of the gamma radiation monitoring in the period 1992 – 2009 showed that the contamination by the radioactive substances was decreasing. The decrease can be described by an equation, which is used for description of kinetic of the first order. The range of the radium 226 activities in bottom sediments from the cross sections of the Ploučnice River was relatively wide. It could be, however, generally concluded that the radium 226 activities were significantly decreasing during the whole monitoring period 1994 – 2009. The results of the monitoring indicate that a control measurement should be repeated in 2014 or after a flood, whose return period would exceed 5 years.

Acknowledgments

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